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by Dr. John K. Brennan, Dr. Linhbao Tran, and Dr. Müge Fermen-Coker

ARL-TR-4753 **March 2009**

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14. ABSTRACT

A critical Army mission is to improve predictive technologies for the development of future weapon systems. Shear initiated reactions are an important aspect of lethality, survivability, and vulnerability considerations, i.e., the increased lethal effects due to shear localization of reactive materials, reactive armor applications, and shear-induced reactions in munitions due to fragment impact. Present computational capabilities in continuum mechanics codes used by Army designers do not possess the capability to properly simulate these events, and therefore, cannot be used effectively to develop advanced weapons concepts. In this report, we discuss the development of a multi-scale framework to simulate and predict shear initiated reactions in energetic and reactive materials. First, we implemented the framework into an Eulerian wave propagation code. Then, using the energy conserving version of the Dissipative Particle Dynamics Method (DPDE) as the mesoscale method, we developed a sub-grid model to incorporate mesoscale output into the continuum level and used an existing localization model at the continuum level.

15. SUBJECT TERMS

Shear initiated reactions, energetic, HE, reactive materials, IM, multi-scale modeling

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1. Objective

The objectives are as follows:

1. Formulate and develop a multi-scale approach for simulating shear initiated reactions that will span from the mesoscale to the full continuum scale, to link the length scale of material heterogeneities to the length scale of the weapon system.
3. Develop models and establish bridging at the various length scales, followed by verification and assessment of the multi-scale approach.

The final product is an improved predictive capability implemented into the Combined Hydro and Radiation Transport Diffusion (CTH) hydrocode developed by Sandia National Laboratories that allows for the prediction and/or simulation of high explosive (HE) and reactive material (RM) reactions when subjected to loads that result in shear localizations.

2. Approach

A schematic of the approach developed is shown in figure 1. It consists of four major steps as follows:

1. Perform the mesoscale modeling.
2. Convert the mesoscale output to input for the reactive burn model.
3. Link the reactive burn model output to the continuum level simulation of shear initiated reactions.
4. Assess the overall approach by implementing it into the continuum mechanics code CTH, and perform simulations for HEs and RMs.

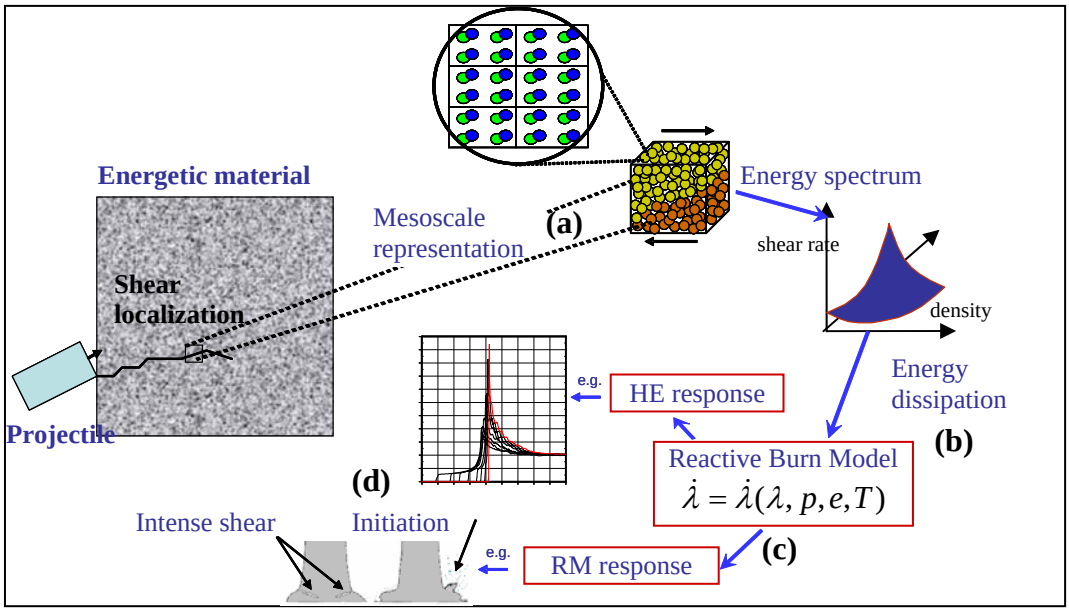

Figure 1. Schematic of multi-scale shear initiation model.

As depicted in figure 1(a), each mesoparticle represents several atomic-level unit cells of the crystalline material. The mesoscale method, Dissipative Particle Dynamics with Energy Conservation (DPDE) (*1*, *2*), provides the input to the reactive burn model. Thermodynamic properties (pressure, density, temperature, and energy) from the mesoscale simulations are generated at various steady-state shear rates, providing look-up data tables for use in the CTH modeling (cf., figure 1(b)). For each strain observed in the computational domain where a shear localization has developed, the state variables and the energy spectrum obtained by the mesoscale model are known at the continuum level. With this approach, the assumptions made in the continuum mechanics code are circumvented. In other words, by incorporating mesoscale level calculations into the continuum mechanics code, more accurate estimates for the thermodynamic state within the localized regions, and consequently, more accurate input to the reactive burn model are obtained.

We developed a sub-grid model to account for energy dissipation, which brings the mesoscale temperature estimates to the continuum level in a relatively cell-size independent manner by calculating a volumetric averaged solution of the extent of reaction at the sub-grid level to represent the continuum extent of reaction (cf. figure 1(c)). We use this extent of reaction in the equation of state to determine the amount of energy release.

The sub-grid calculations are performed for cells that are identified to contain shear bands at the continuum level. A numerical framework for nucleating and propagating shear band localizations already exists in CTH (*3*–*5*). When a set of nucleation criteria is satisfied, a shear band is formed by introducing Lagrangian particles, which conform to local planes of maximum shear as they propagate in three-dimensional space until a set of growth criteria are no longer satisfied along the points defining its boundary. We use this framework to track shear localizations, and we

applied our multi-scale approach at these locations to simulate the shear initiation of energetic and reactive materials (cf. figure 1(d)). We then implement the approach into CTH and conduct simulations for TNT to verify the algorithm. The details of the approach follow.

The energy conserving version of the dissipative particle dynamics method (DPDE) is a particle-based mesoscale method that simulates the hydrodynamic behavior of materials, conserving both momentum and energy while allowing the mesoparticles to exchange both viscous and thermal energy (*1*,*2*). In the DPDE method, the changes with respect to time t in position $\{\mathbf{r}_i\}$, momentum $\{\mathbf{p}_i\}$, and internal energy $\{u_i\}$ of the ith particle with mass m_i due to the interaction with the jth particle can be expressed as

$$\dot{\mathbf{r}}_i = \frac{\mathbf{p}_i}{m_i} \tag{1}$$

$$\dot{\mathbf{p}}_i = \mathbf{F}_{ij}^C + \mathbf{F}_i^{ext} + \mathbf{F}_{ij}^D + \mathbf{F}_{ij}^R \tag{2}$$

$$\dot{u}_i = -\frac{1}{2m_i}\left(\mathbf{p}_i - \mathbf{p}_j\right)\cdot\left(\mathbf{F}_{ij}^D + \mathbf{F}_{ij}^R\right) + \dot{q}_{ij}^D + \dot{q}_{ij}^R \tag{3}$$

with the requirement that $\dot{q}_{ij}^D = -\dot{q}_{ji}^D$ and $\dot{q}_{ij}^R = -\dot{q}_{ji}^R$, where $\dot{q}_{ij}^D$ and $\dot{q}_{ij}^R$ are the dissipative and random mesoscopic heat flows, respectively, and where the dot notation denotes time-differentiation. $\mathbf{F}_{ij}^C$ and $\mathbf{F}_i^{ext}$ are the forces due to the conservative interactions, while $\mathbf{F}_{ij}^D$ and $\mathbf{F}_{ij}^R$ are the dissipative and random forces, respectively. A particle temperature, θ_i, is defined as $\theta_i \equiv \partial u_i / \partial s_i$, where s_i is defined as a mesoscopic entropy. In general, this microscopic state law, or *mesoparticle equation of state*, $\theta = \theta(u)$, can be determined from molecular simulations, first-principle calculations, or available experimental data.

We induced steady planar shear flow by means of the Lees-Edwards boundary conditions (*6*), in which the simulation box and its images centered at $(x, y) = (\pm L, 0), (\pm 2L, 0), \ldots$, are taken to be stationary, while boxes in the layer above, $(x, y) = (0, L), (\pm L, L), (\pm 2L, L), \ldots$, are moving at a speed ϖL in the positive x direction, where ϖ is the shear rate. Boxes in the layer below, $(x, y) = (0, -L), (\pm L, -L), (\pm 2L, -L), \ldots$, move at a speed ϖL in the negative x direction. A system under such conditions is subjected to a uniform steady shear in the xy plane.

Next we parameterized a mesoparticle potential for TNT by fitting to the Mie-Grüneisen equation of state at several state points. Mesoparticles interact through a pairwise additive third-order Rydberg potential given as

$$\psi_R\left(r_{ij}\right) = \begin{cases} -\varepsilon\left(1 + d + \alpha d^3\right)e^{-d} & \text{for } r_{ij} \le r_{cut} \\ 0 & \text{for } r_{ij} > r_{cut} \end{cases}, \tag{4}$$

where, r_{ij} is the separation distance between particle *i* and *j*, and $d = \sigma\left(\frac{r_{ij}}{r_0} - 1\right)$. As a first approximation, we performed fitting by specifying r_0=5.07 Å, σ=7.90, α=0.185, and r_{cut}=15.0 Å, and subsequently determining ε=1.612x10^{-20} J. A mesoparticle was chosen to represent a single TNT molecule.

From statistical thermodynamics, the internal particle energy of an isolated nonlinear polyatomic molecule containing *n* number of atoms can be expressed as the sum of the translational, rotational, vibrational, and electronic contributions, respectively, so that

$$\frac{u}{k_B T} = \frac{3}{2} + \frac{3}{2} + \sum_{j=1}^{3n-6}\left(\frac{\Theta_{vib,j}}{2T} + \frac{\Theta_{vib,j}/T}{e^{\Theta_{vib,j}/T} - 1}\right) - \frac{D_e}{k_B T}, \tag{5}$$

where $\Theta_{vib,j}$ is the characteristic vibrational temperature, and D_e is the depth of the ground electronic state potential well (*7*), both of which can be determined from quantum mechanics calculations or taken from thermodynamic tables (*8*) when available, and where k_B is the Boltzmann constant, *T* is the temperature. As a first approximation for this study, we formulate equation 5 based upon the classical limit (relatively high temperature) (*7*), so that

$$\frac{u}{k_B T} = \frac{3}{2} + \frac{3}{2} + \left(3(21) - 6\right) = 60, \tag{6}$$

since each TNT molecule contains 21 atoms and is represented by a single mesoparticle. By replacing *T* with the internal particle temperature θ_i, we arrive at the equation of state for particle *i*

$$\theta_i = \frac{u_i}{60 k_B}. \tag{7}$$

A sub-grid model is developed to account for the dissipation of energy and allow for delayed reactions. The initiation phenomenon in heterogeneous energetic materials can occur when the material is subjected to impulses such as shock waves. Under sufficiently strong shock conditions, these shock waves can evolve into self-sustaining detonation waves. This process, shock-to-detonation transition (SDT), is fairly well understood at the phenomenological level and is based on the theory of hot spot formation (such as void collapse, visco-plastic heating, shear band, frictional heating, etc.). However for conditions such that the SDT process does not occur, the initiation of HE leading to explosion can take place if the material has been sufficiently confined. Such analysis was shown by Frey (*9*), whose study indicated that both pressure and shear rate were important parameters in controlling runaway explosion. Another study showed that a shear banding mechanism could provide the large ignited surface area,

which is believed to be necessary to explain shock initiation (*1*). The long delay between the time at impact and explosion can typically be hundreds of milliseconds; whereas, for the SDT process, it is on the order of microseconds. Such dominant physical features suggest energy competing processes at the micro-scale level. Therefore, at hot spot locations, e.g., shear surfaces, the possibility of initiation hinges on a balance (or lack thereof) between energy producing mechanisms (visco-plastic work, shear localization, chemical reaction, etc.) and the rate at which the energy is transported away from the zone. Energy generated from shear localization within the narrow region of the shear band width, which is on the order of micrometers, can be much higher than the bulk energy in the typical cell size of continuum simulations, which is on the order of millimeters. The large temperature gradient within a continuum cell supports the need to account for thermal diffusion. Figure 2 illustrates temperature profiles, including energy release due to reaction, as time progresses. This concept forms the basis of our sub-grid model.

Within each sheared continuum cell, we assume that the shear band surface is located at the mid-plane (figure 2). Such a geometric assumption leads to a simple and tractable solution at the sub-grid level. Each sheared cell is subdivided into intervals, and time dependent equations of temperature and species are described as follow:

$$\rho C \frac{\partial T}{\partial t} = k \frac{\partial^2 T}{\partial x^2} + \Delta H \dot{m}''' \tag{8}$$

$$\rho \frac{\partial \lambda_{sg}}{\partial t} = \rho D \frac{\partial^2 \lambda_{sg}}{\partial x^2} - \dot{m}''', \tag{9}$$

where, λ_{sg} is the extent of reaction at the sub-grid level, heat of reaction is ΔH, diffusion coefficient is D, thermal conductivity is k, and the volumetric mass production rate, $\dot{m}'''$, is governed by an Arrhenius-type reaction:

$$\dot{m}''' = A\rho\left(1-\lambda_{sg}\right)\exp\left(-T_a \big/ T\right), \tag{10}$$

where A is the frequency and T_a is the activation temperature. The species diffusion term is assumed negligible in the current study. Solutions for the above equations are integrated using an implicit method with the initial conditions:

$$\begin{aligned} T(x,0) &= T_{meso}, \qquad 0 \le x \le w_{sb} \\ T(x,0) &= T_{cell}, \qquad w_{sb} < x \le \Delta L \big/ 2 \end{aligned}, \tag{11}$$

where the shear band temperature, T_{meso}, is obtained from mesoscale simulations of a shear banding process, T_{cell} is the continuum cell temperature, and w_{sb} is the shear band width. Boundary conditions are given as

$$\left.\frac{\partial T}{\partial x}\right|_{x=0} = 0 \text{ and } T\left(x = \Delta L/2\right) = T_{cell} \tag{12}$$

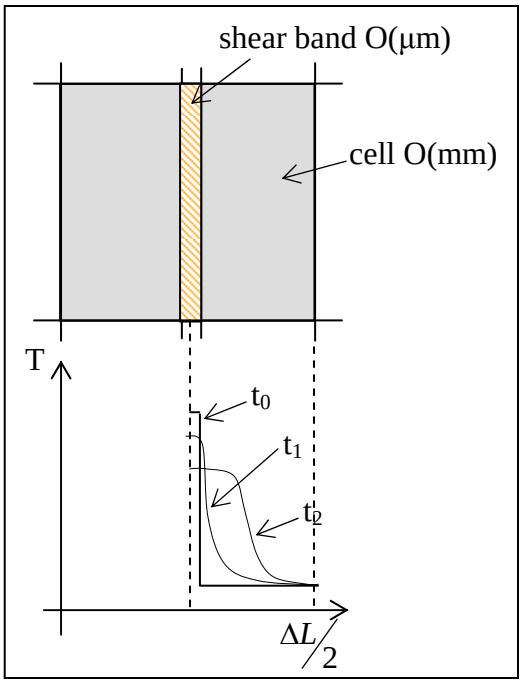

Figure 2. Sheared cell and its temperature profile.

With each sheared cell, a volumetric averaged solution of the extent of reaction at the sub-grid level is transferred to the continuum level as the continuum extent-of-reaction. This extent-of-reaction is used in the equation of state to determine the amount of energy release. Such an approach ensures consistency in our internal energy calculation.

3. Results

We generated the mesoscale look-up table for a range of densities (1.65, 1.70, 1.80, 1.90, and 2.00 g/cm^3) and three shear rates (50, 75, and 100 s^{-1}), calculating both pressure and temperature. Figure 3(a) shows a sample of the data found in the look-up table. Figure 3(b) shows a sample energy spectrum determined by the mesoscale model depicting the internal temperature spike due to shearing.

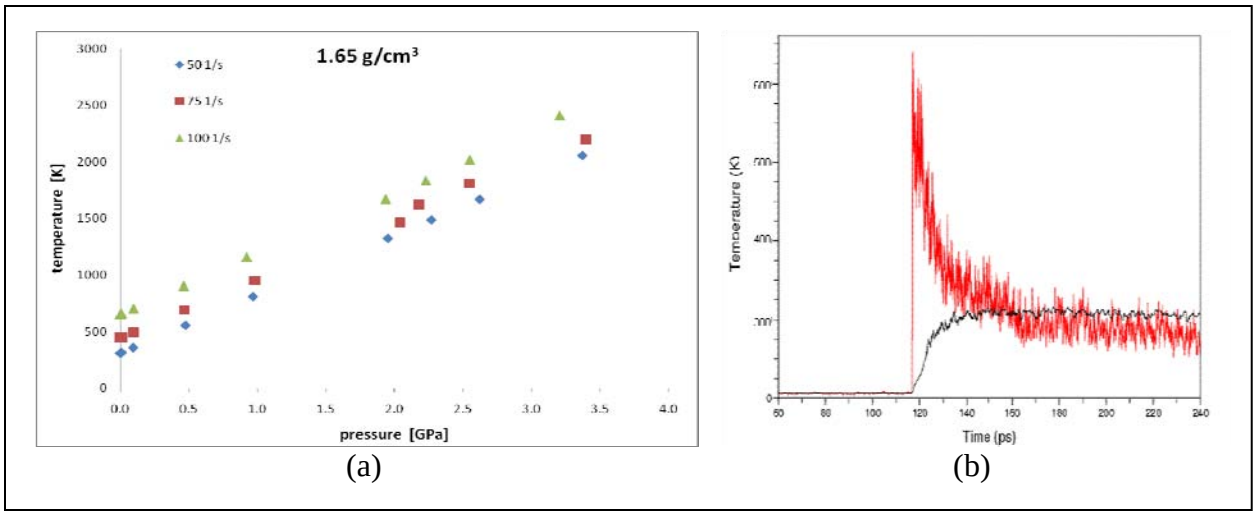

Figure 3. (a) Mesoscale temperatures calculated as a function of pressure and shear rate, and (b) temporal variations of internal (black) and kinetic (red) temperatures after the system has been sheared in the mesoscale simulation.

Using TNT parameters, we subjected a block of material to pure shear by imposing a velocity on the lower half of the block to verify the newly implemented approach in CTH, as shown in figure 4. The simulation represents a hypothetical configuration to simply conduct a numerical test and demonstrate the depiction of a reaction that previously was unattainable.

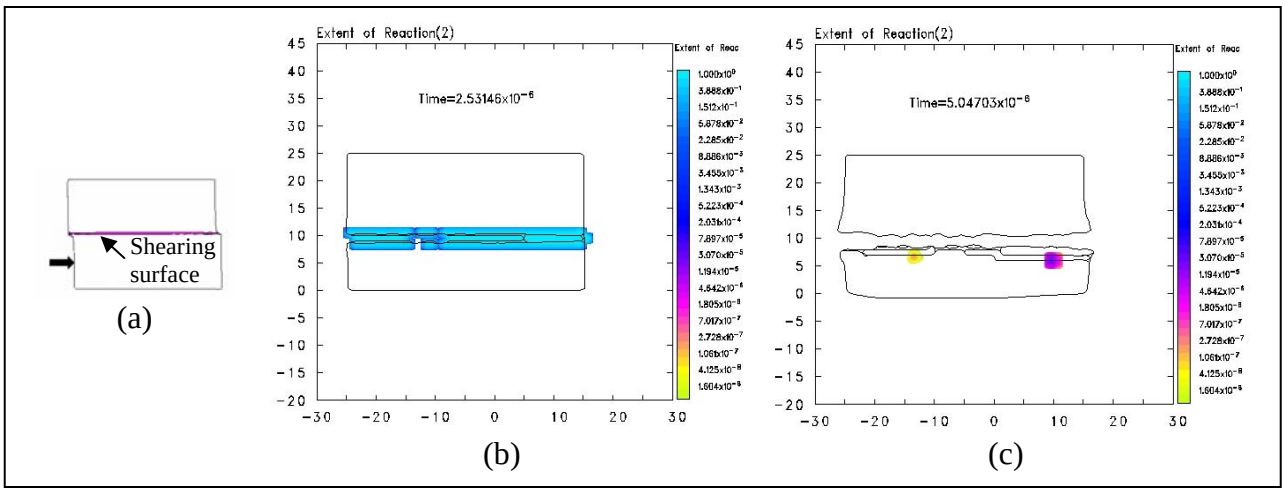

Figure 4. Example problem to verify a new multi-scale approach: (a) sliding half a block of material, (b) extent of reaction at 2.5 μs, and (c) extent of reaction at 5 μs.

4. Conclusions

In this effort, we developed a multi-scale approach for simulating shear initiated reactions that span from the molecular scale to the mesoscale (length scale of material heterogeneities) to the full continuum scale (length scale of the weapon system). We then constructed a framework based on our approach, implemented it into the CTH hydrocode developed by Sandia National Laboratory, and demonstrated that the new approach allows predictions not previously possible for energetic materials when subjected to loads that result in shear localizations.

This computational tool provides a novel modeling capability that opens previously unavailable avenues by bridging the gaps between multiple scales. This capability enables improved predictions towards designing armor and anti-armor devices. It also supports the development of concepts for enhanced survivability and lethality, primarily in the areas of insensitive munitions and reactive armor, and of novel concepts and designs using RMs.

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6. Transitions

Transition of the multi-scale model to Sandia is planned and will be executed following the completion of the effort in fiscal year 2009 (FY09).

List of Symbols, Abbreviations, and Acronyms

CTH	Combined Hydro and Radiation Transport Diffusion
DPDE	Dissipative Particle Dynamics with Energy Conservation
FY09	fiscal year 2009
HE	high explosive
RM	reactive material
SDT	shock-to-detonation transition

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